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PERMEATION OF TRITIATED WATER VAPOUR THROUGH PLASTIC PACKAGING MATERIAL

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ABSTRACT

A new respirator for tritiated water vapour removal has been selected for service at Ontario Hydro nuclear generating stations. The respirator utilizes two cartridges saturated with deionized water (about 37% by weight). Each pair of cartridges is received sealed in a 4-layer, 5.0 mil thick plastic pouch to prevent the egress of moisture prior to use. The permeation of HTO vapour through the plastic packaging material during storage in atmospheres containing chronic amounts of tritiated moisture (<100 MPCa) is a concern to station radiation control personnel. A passive tritiated water vapour permeation experiment with a representative section of the plastic packaging was therefore carried out. An HTO permeation rate of 0.0133 nCi cm^{-2} day^{-1} was determined. This means that a typical sealed package of cartridges (500 cm^2 surface area) exposed for 25 days to a 100 MPCa HTO atmosphere, could lead to a breathing air equivalent HTO concentration of about 0.0051 MPCa. This is negligible compared to the concentration of HTO (up to 100 MPCa) that the operator could experience when working at an Ontario Hydro NGS.

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1.0 INTRODUCTION

A replacement for the Stephenson respirator, used for protection against low-level tritiated water vapour (<100 MPCa) at Ontario Hydro NGS sites, has been selected. The new respirator is a modified version of the Mine Safety Appliance (MSA) Company Comfo-II dual cartridge respirator. Each of the two cartridges contains fine activated charcoal saturated with deionized water (about 37% by weight). After manufacture, each cartridge pair is placed into a polyethylene/polypropylene/aluminum foil laminated pouch and heat sealed shut to prevent the egress of moisture during storage.

After sealing the respirator cartridges into the pouch, a small amount of moisture from the cartridges was found to wet the inside of the pouch.

At Ontario Hydro NGS sites, a sealed package of respirator cartridges could be stored for extended periods of time in locations where chronic exposures to levels of tritiated moisture up to 100 MPCa is possible. This is a matter of concern. As a result, Ontario Hydro NGS safety officers have asked for direct evidence that the permeation of tritiated moisture through the packaging material during storage in areas that contain elevated tritiated water vapour concentrations is negligible.

A passive permeation experiment simulating field exposure conditions in a tritiated water vapour environment has been successfully completed. This report presents a description of the experiment in Sections 2.0, the results and discussion in Sections 3.0 and the conclusions and recommendations in Section 4.0.

2.0 EXPERIMENTAL

2.1 Test Cell

A passive permeation cell was designed and built, see Figure 1. Three 50 mm ID KF-type stainless steel flanged fittings, one with the tube-end welded shut with a stainless steel blank to form the lower chamber and the other two KF-type fittings butt welded together to form the upper chamber were used. A removable sealing plate, two sealing 'O' rings and two sealing KF-flange clamps completed the test cell. The sample of packaging material to be tested was sealed between the upper and lower sections of the test cell with the help of a sealing 'O' ring and a KF-flange clamp.

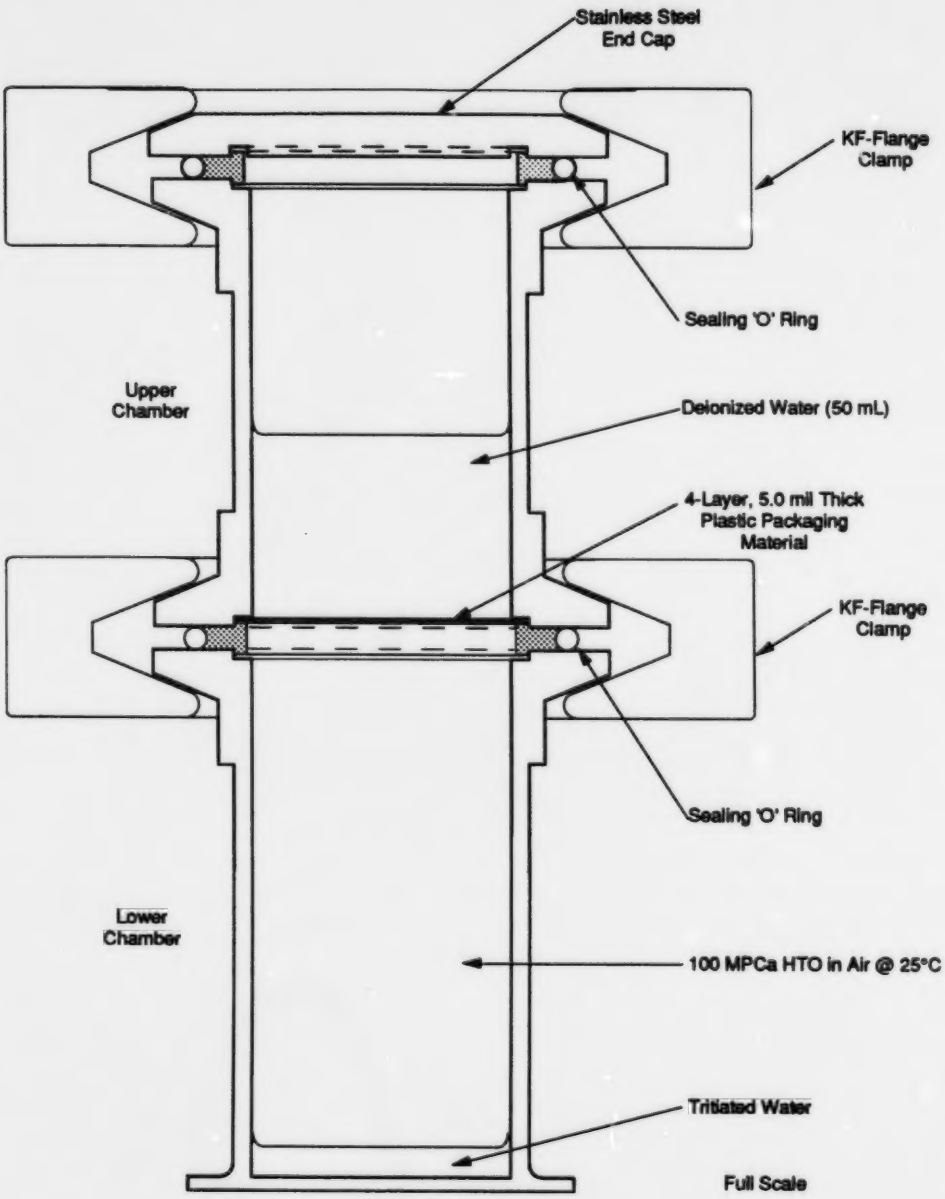

FIGURE 1
SCHEMATIC OF STAINLESS STEEL TEST CELL FOR THE STUDY OF PASSIVE TRITIATED WATER VAPOUR PERMEATION OF PLASTIC PACKAGING MATERIALS

2.2 Test Material

The material used to package pairs of MSA GMT respirator cartridges conformed to the US Military specification MIL-B-131-H (17 February, 1987)/1/. The specification established the minimum requirements for a heat-sealable, greaseproof, flexible barrier material having a low water vapour transmission characteristic when used in packaging for the US Military. The classification of the packaging material is: Type-I - non-flame resistant; Class 1 - plastic, non-woven backing. Barrier materials can be constructed of one or more plies in any manner which ensures compliance with the performance requirement of the specification. Water vapour transmission rate (WVTR) after flexing/aging at room temperature should not exceed 0.02 g/100 sq in/day (0.000031 g cm^{-2} day^{-1}). The packaging material should not delaminate during prolonged exposure to moisture.

The sample of packaging material used in the passive permeation experiment described in this report was cut from an actual package which contained a pair of MSA GMT respirator cartridges that were tested for tritiated moisture removal performance. The packaging material used to seal the recent shipment of MSA GMT respirator cartridges was produced by Bell Fiber of Columbus, Georgia. The material has a nominal thickness of 5.0 mil and it is four ply in construction. The layer exposed to the ambient air is polypropylene followed by a polyethylene bonding layer, a layer of aluminum foil and finished with another polyethylene layer that is in contact with the cartridges.

2.3 Test Procedure and Sampling

The test cell was cleaned and dried and leak tested prior to use. The tritiated water solution (50 µCi/mL) was added to the lower part of the cell. The resulting tritium in air concentration would be approximately 100 MPCa (1000 µCi/M^3) at room temperature (25°C) when the cell was sealed.

The sample of plastic packaging material was placed onto the 'O' ring seated on the flange of the lower portion of the test cell. A flange of the upper portion of the test cell was carefully seated onto the test material that was sitting on the 'O' ring. A KF-flange clamp was attached and turned down until tight.

To simulate the wetting of the interior layer of the sealed package, 50 mL of deionized water was poured directly onto the packaging material in the upper portion of the test cell. The

upper portion of the test cell was sealed with a stainless steel blank, 'O' ring seal and a KF-flange clamp.

The test cell was allowed to come to temperature equilibrium. The first sample of the water layer was taken after 3 h. A second sample was taken after 6 h. Subsequent samples were taken twice a day where possible over the period of a month. Prior to taking a sample, the test cell was gently swirled to ensure the deionized water layer was well mixed. Each 1 mL water sample was added to 15 mL of cocktail (Monophase 'S' scintillation fluid) and counted on a Packard scintillation counter for five minutes. With each sample from the test cell, a deionized water blank was also counted. At the end of the test period a 1 mL sample of the tritiated water layer was taken and counted to confirm the source strength.

3.0 RESULTS AND DISCUSSION

A passive tritiated water vapour permeation test on a 4.0 cm^2 piece of the multilayer packaging material was successfully completed. Over 600 h of continuous exposure to a 100 MPCa equivalent HTO atmosphere was monitored with 1 mL grab samples of the water layer. A total of 32 samples were taken from the water layer and counted for the HTO content. The corrected results are presented in Table I and displayed in Figure 2.

For the experiment described in this report steady state permeation can be assumed. The total amount of HTO diffusing, therefore, varies linearly with time. The relevant equations have been derived in an earlier report /2/. The permeation rate can be derived from the slope of the total amount diffusing versus time relationship at steady state. The rate of permeation of HTO from the saturated 100 MPCa HTO atmosphere in the lower chamber through the 4.0 cm^2 piece of 4-layer, 5.0 mil thick packaging material into the water layer was estimated by linear regression and found to be 0.0133 nCi cm^{-2} day^{-1}.

For comparison, the US Military specification, to which the material tested must conform, allows a maximum water permeation of 31.0 µg cm^{-2} day^{-1}. For the passive test, the HTO concentration in the lower chamber was 46.2 µCi g^{-1}. Therefore a maximum HTO penetration rate for the packaging material could be estimated and it was found to be 0.143 nCi cm^{-2} day^{-1}. The measured HTO permeation rate for the packaging material was less than 10% of the maximum estimated rate allowed for the packaging material.

TABLE I

TRITIATED WATER VAPOUR PERMEATION THROUGH RESPIRATOR CARTRIDGE PACKAGING MATERIAL - EXPERIMENTAL RESULTS

Total Exposure (hours)	Total HTO Permeation ($Ci \times 10^{-9}$)	Total HTO Permeation per cm^2 of Test Material ($Ci \times 10^{-9}$)
0.0	0.0	0.0
3.1	0.085	0.0213
6.1	0.0	0.0
23.9	0.098	0.0245
30.5	0.0	0.0
95.7	0.093	0.0233
103.2	0.183	0.0458
119.4	0.151	0.0377
126.7	0.305	0.0763
143.5	0.396	0.0990
150.8	0.294	0.0735
166.4	0.445	0.1113
287.7	0.622	0.1555
294.9	0.574	0.1435
311.4	0.664	0.1660
318.8	0.698	0.1745
335.4	0.603	0.1508
342.8	0.684	0.1710
359.3	0.693	0.1733
366.8	0.698	0.1745
413.4	1.210	0.3025
438.1	0.988	0.2470
454.3	1.074	0.2685
461.7	0.911	0.2278
478.4	1.096	0.2740
485.4	0.934	0.2335
502.9	0.995	0.2490
509.3	1.115	0.2790
526.9	1.120	0.2800
533.7	1.146	0.2865
598.1	1.255	0.3138
606.1	1.507	0.3768
625.6	1.394	0.3485

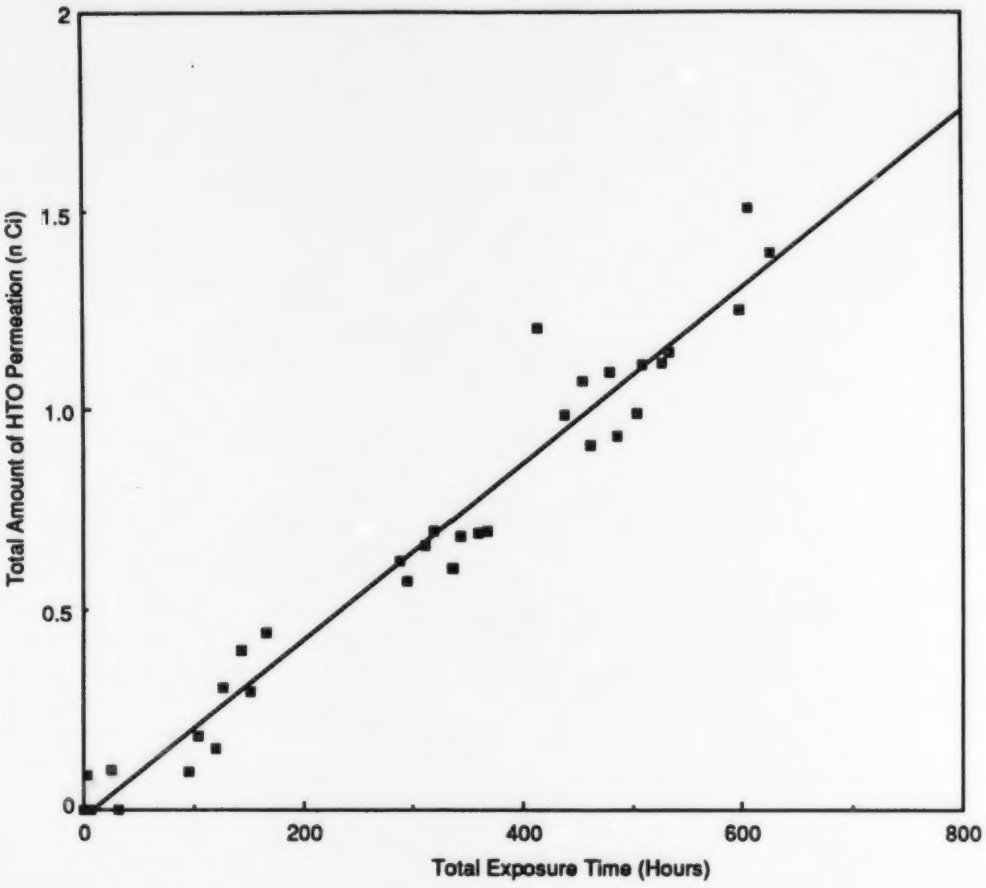

FIGURE 2
HTO PERMEATION THROUGH 4-LAYER, 5.0 mil THICK PLASTIC PACKAGING AS A FUNCTION OF TOTAL EXPOSURE TIME

Since the multilayer packaging material was composed of two layers of high density polyethylene (HDPE), a comparison to an equivalent 5.0 mil thick HDPE material can be made with data obtained in an earlier study/3/. From the earlier data and using the 5.0 mil thickness and unit area (1 cm^2) the rate of HTO penetration was estimated to be 0.969 nCi cm^{-2} day^{-1}. The rate of HTO permeation measured for the 4-layer, 5.0 mil thick plastic material evaluated in this study was less than 1.4% of the HTO permeation value estimated for the equivalent 5.0 mil thick HDPE.

To attempt to put the permeation rate values on a more understandable basis the following conditions were specified. A pouch containing two respirator cartridges was assumed to have 500 cm^2 of surface area available for tritiated water vapour permeation. The external atmosphere was assumed to have a constant HTO concentration of 100 MPCa (1000 $\mu Ci/M^3$) surrounding the packaged canister for a total of 25 days. Each respirator cartridge was estimated to have about 100 g of charcoal containing 37% by weight of untritiated water. The total available water in each pouch was then estimated to be about 75 ml. The total permeation of HTO into the package as estimated from the experimental data would be about 166 nCi. Assuming equal distribution, the HTO concentration in the pouch was calculated to be about 2.22 nCi/mL. At 25°C and saturated moisture conditions, the breathing air from the cartridges would have an equivalent HTO concentration of 0.0051 MPCa. This is negligible when compared to the high concentration (up to 100 MPCa) of HTO to which the user could subject the respirator when breathing HTO contaminated air.

Similarly, for the worst case, the maximum allowed permeation rate (0.143 nCi cm^{-2} day^{-1}) yields a breathing air HTO concentration at 25°C of 0.055 MPCa when the cartridges are used in a respirator. This again is negligible when compared to the high HTO exposure that the user would subject the respirator.

4.0 CONCLUSIONS AND RECOMMENDATIONS

The following conclusions result directly from the work performed:

- The multilayer plastic material used to package the pairs of MSA GMT respirator cartridges performed very well in preventing tritiated moisture ingress.

- The HTO permeation rate for the 5.0 mil thick, 4-layer plastic packaging material was estimated to be 0.0133 nCi cm^{-2} day^{-1}.

- The breathing zone HTO concentration from respirator cartridges stored in the plastic package for a 25 day period in a 100 MPCa HTO atmosphere at 25°C was estimated to be <0.01 MPCa.

It is recommended that the 4-layer, 5.0 mil thick packaging material continue to be used to seal the respirator cartridges. The material was found to perform very well in tritiated atmospheres up to 100 MPCa. It is also recommended that if a package of respirator cartridges is found to be perforated prior to use then the cartridges should not be used. A new sealed package of respirator cartridges should be obtained and used.

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